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TOWARD AN UNDERSTANDING OF THE MECHANICAL MIXED ALKALI EFFECT I--ETC(U)

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Toward an Understanding of the Mechanical Mixed Alkali Effect in Glasses

by

Costas H. Tzinis and William M. Risen, Jr.

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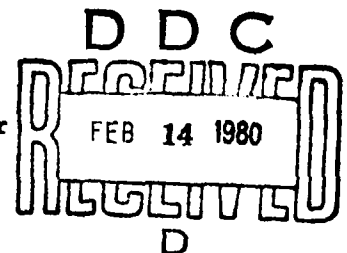
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The mechanical properties of mixed alkali glasses have been analyzed in order to determine their common features, including systematic variations with composition, and to define clearly the phenomena that must be explained. New assignments of the mechanical loss peaks are made, and approaches to developing a cooperative theory for the phenomena, especially the compositional variation of the peak temperatures and loss tangents, are presented and discussed.		

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Toward an Understanding of the Mechanical Mixed Alkali Effect in Glasses

Introduction

Mixed alkali glasses, systems with the overall composition $xM_2O(1-x)M'_2O \cdot ApO_q$ (where M and M' are alkali metals, and ApO_q is a network former) exhibit various dependencies on the moles fraction, x, of alkali (1). Major deviations from linearity are observed for mechanical relaxation and for properties related to ionic (mass) transport, such as conductivity and electrical relaxation. Minor deviations are found for density, molar volume, refractive index and thermal expansion. As an illustration of the significance of such deviations, the conductivity of a mid member glass in a series ($x = 0.5$) can be as much as five orders of magnitude less than that of the end member ($x = 0.0$ or 1.0) glass (2). The dramatic changes that occur when a second alkali is introduced are usually referred to collectively as the mixed alkali effect. This effect has recently been explained for the case of ionic conductivity on the basis of the cooperativity of cation motion in the glass network (3). On the other hand, the mechanical properties of mixed alkali glasses are not well understood. Part of the problem is to define clearly what phenomena a theoretical interpretation must explain, because many observations have been made on dissimilar glasses and relatively few systematically varied series have been studied.

In this report we focus on analyzing the data from studies of the mechanical properties of mixed alkali glasses. They involve measuring the energy loss or dissipation (usually labelled $\tan \delta$ or Q^{-1}) as a function of temperature at a given frequency. Such measurements will be referred to as the mechanical spectrum. A spectrum typically is determined between about -200°C and close to the glass transition temperature at frequencies ranging from about 0.01 to 5×10^6 Hz. To cover this frequency range the torsion pendulum is used in the 0.1 to several Hertz range, the fluxural beam method in the kilohertz

range, and the pulse-echo method in the megahertz range (4). It would be more useful, of course, to have the energy loss as a function of frequency at constant temperature, but this commonly is not available because of experimental limitations.

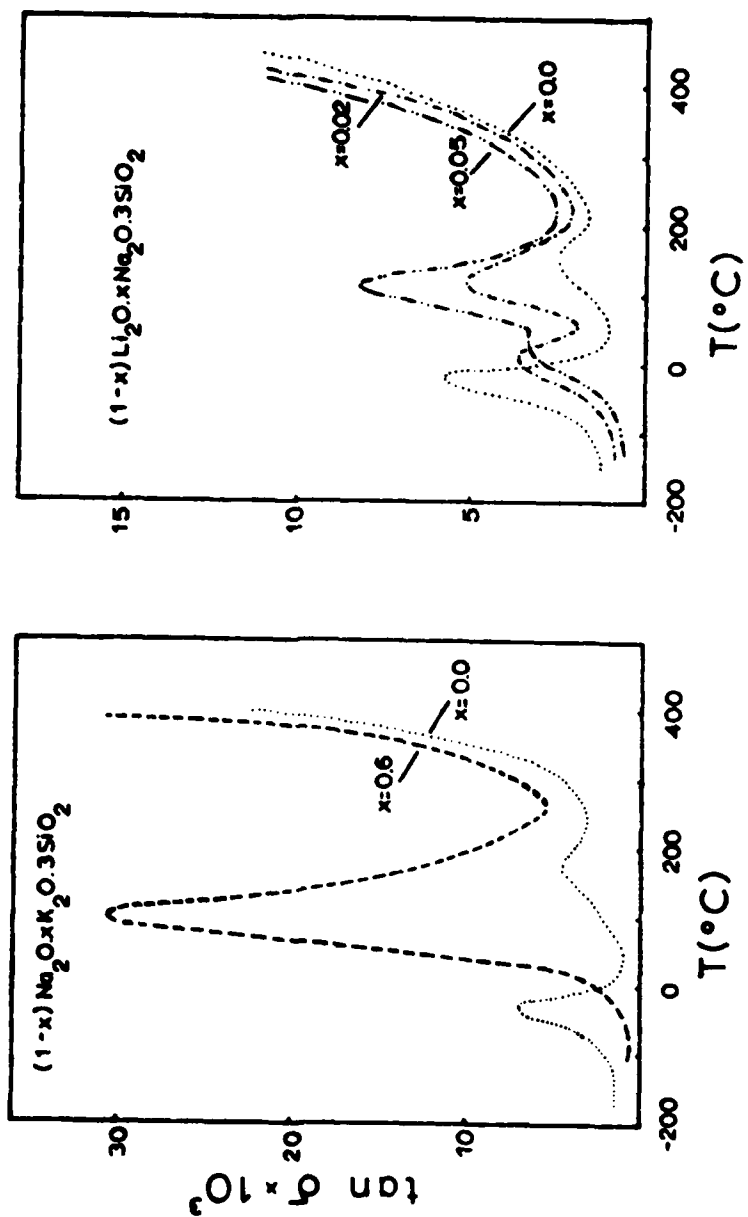
The main mechanical effect of the introduction of a second alkali into an ionic oxide glass is that at least one new peak appears and grows to dominate the mechanical loss spectrum. Data on several mixed alkali systems are presented in Figure 1, to illustrate this effect.

Although the introduction of the second alkali clearly has a pronounced effect on the mechanical properties, the effect is not well understood, and there is considerable confusion in the literature as to its origin. Indeed, there has not been a systematic attempt to interpret and analyze all existing data so that an understanding of the mechanism(s) responsible for the inelasticity, based on the inherent cooperativity of motion in a tightly coupled random network, can be developed. In this report we attempt to analyze the existing data on the "mechanical mixed alkali effect", define clearly what phenomena a theoretical interpretation must explain, and to suggest some directions for its development.

It is important to note at the outset that no connection between mechanical loss and electrical conductivity has yet been shown. Although it has been proposed that the higher temperature peak (HTP) is controlled by the rate of diffusion of the slower moving cation (5), it is surprising that no such general connection has been suggested since both ionic conductivity and mechanical properties clearly involve cation motion in ionic oxide networks. It also is useful to recall, as previously pointed out in connection with the mixed alkali effect on conductivity (3), that any motion relevant to a relaxation process is inherently cooperative. This requires that all cations, in fact all atoms, participate in

Figure 1

Mechanical Loss Spectra of Several Mixed Alkali Trisilicate Glass Systems (1,6)



a relaxation process, if only through their involvement in the vibrational relaxation accompanying the process. Although certain inherently collective motions, such as bond vibrations, can usefully be considered to be localized to the motion of a few atoms, motions at frequencies lower than about 10^{10} Hz must be considered cooperative. Clearly the importance of this cooperativity varies with the process, but for large deformations of a system and for measurements at frequencies lower than fundamental vibrational and rotational ones, many coupled fundamental processes occur and must be averaged appropriately.

Analysis of Data

The available data are in the form of mechanical loss tangent ($\tan\delta$ or Q^{-1}) as a function of temperature at a given frequency for a given composition. If the standard inelastic model for a thermally activated process is assumed to apply (5), the apparent activation energy, E_a , can be calculated from the frequency dependence of the mechanical resonance. For this model it can be shown that

$$\tan\delta = \frac{\delta M}{M_r} \frac{\omega\tau}{1+(\omega\tau)^2} \quad (1)$$

where δM , M_r , ω , and τ , are, respectively, the difference between the relaxed and unrelaxed mechanical modulus, the relaxed modulus, the experimental measurement frequency, and the relaxation time of the measured process. The relaxation time, τ , for a thermally activated process is given generally by

$$\tau = \tau'_0 \exp \frac{\Delta G}{RT} = \tau'_0 \exp \frac{E_a}{RT} \quad (2)$$

The last part of Eqn. 2 is commonly used to analyze experimental data. Since mechanical resonance demands that $\omega\tau = 1$, a peak appears in the $\tan\delta$ vs T spectrum when

$$\omega\tau'_0 \exp \frac{E_a}{RT} = 1 \quad (3)$$

If the resonance temperatures T_{R_1} and T_{R_2} are known at two different circular frequencies, ω_1 and ω_2 , the apparent activation energy is given by

$$E_a = \frac{-R \ln(\omega_1/\omega_2)}{\frac{1}{T_{R_1}} - \frac{1}{T_{R_2}}} = \frac{-R \ln(f_1/f_2)}{\frac{1}{T_{R_1}} - \frac{1}{T_{R_2}}} \quad (4)$$

where the f_i are the measuring frequencies. In order to derive Eqn. (4) τ_0 must be assumed to be temperature-independent.

Since these methods do not depend on the mechanism of the relaxation, the same analysis applies to both the peak present in the single alkali glasses and the peak present only in the mixed alkali glasses. These peaks often are referred to in the literature as the single alkali (lower temperature) peak and the mixed alkali (higher temperature) peak (6). Since this nomenclature is more confusing than helpful, we will call them LTP and HTP, respectively.

In order to introduce the composition dependence into the relaxation time, we recall that (3)

$$\tau(x) = \tau'_0(x) \exp \frac{\Delta G^*(x)}{RT} \quad (5)$$

where the entire composition dependence is incorporated into the activation free energy, $\Delta G(x)$, and τ'_0 is a constant. In terms of activation enthalpy and entropy, Eqn. (5) is written as

$$\tau(x) = \tau'_0 \exp \frac{-\Delta S^*(x)}{R} \exp \frac{\Delta H^*(x)}{RT} \quad (6)$$

We can compute the individual activation parameters ΔH and ΔS from the frequency dependence of the mechanical resonance, even though it is more complex than in the case where the preexponential factor is assumed to have no composition dependence, from the expressions:

$$\Delta H(x) = \frac{R \ln(\omega_1^{T_{R_2}}/\omega_2^{T_{R_1}})}{\frac{1}{T_{R_2}} - \frac{1}{T_{R_1}}}$$

and $\Delta S(x) = R \left[\ln \frac{\omega_1 h}{T_{R_1} k} + \frac{T_{R_2} (\ln \omega_1^{T_{R_2}}/\omega_2^{T_{R_1}})}{T_{R_1} - T_{R_2}} \right] \quad (7)$

The apparent activation energy, E_a , calculated from Eqn. (2) and the activation enthalpy, ΔH , from Eqn. (7) do not differ by more than the typical experimental error ($\pm 1 \text{ kcal mol}^{-1}$).

The experimental data on those mixed alkali glass series for which enough information is available to evaluate the variation in mechanical behavior as a function of composition are included in this report. Data have been excluded in those cases where the glasses are subject to significant changes if extreme precautions are not taken. Thus, the metaphosphate glasses ($xM_2O(1-x)M'_2O \cdot P_2O_5$) have been excluded because these glasses change significantly when water is present at the parts per million level. Plots of all the valid data known to us in the form of peak temperatures at mechanical resonance, and internal friction magnitudes ($\tan \delta$), and derived in the form of activation energies, E_a , and preexponential factors, τ_0 , versus alkali mole fraction x are presented in Figures 2 and 3.

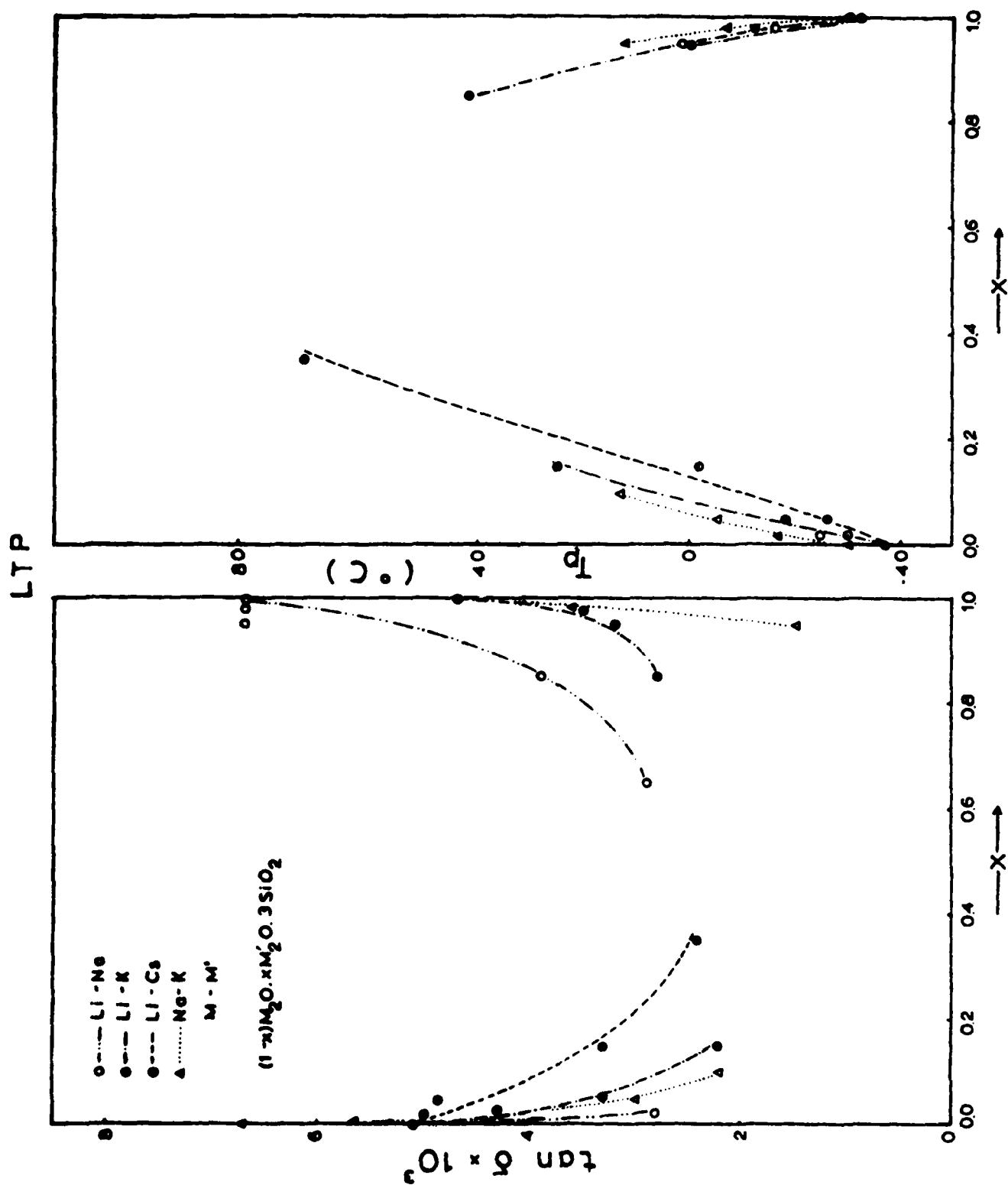
Interpretation of Data

The analysis of the known data makes it possible to see that there are common systematic variations of the mechanical spectral features. We focus first on the main characteristics of the two peaks, the peak temperature, $T_p(x)$, and the magnitude of the loss tangent at the peak, $\tan \delta(x)$.

A careful inspection of the mechanical spectra reveals that the LTP always shows up at a higher temperature (T_p) in mixed alkali glasses than it does in either of the component (end member) single alkali glasses of the series. The activation energy for it varies linearly with T_p , as shown by Eqn. (3), at constant ω and τ_0 . Thus, the composition dependence of the activation energy (for the compositions for which the LTP is clearly distinguishable from the background), exhibits a maximum near $x = 0.5$. Both the magnitude and composition dependence

Figure 2a

Mechanical Loss Peak Temperatures (T_p) and Loss Tangent ($\tan \delta$) for the Low Temperature Peak of Several Mixed Alkali Glass Systems



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Figure 2b

Mechanical Loss Peak Temperatures (T_p) and Loss Tangent ($\tan \delta$) for the High Temperature Peak of Several Mixed Alkali Glass Systems

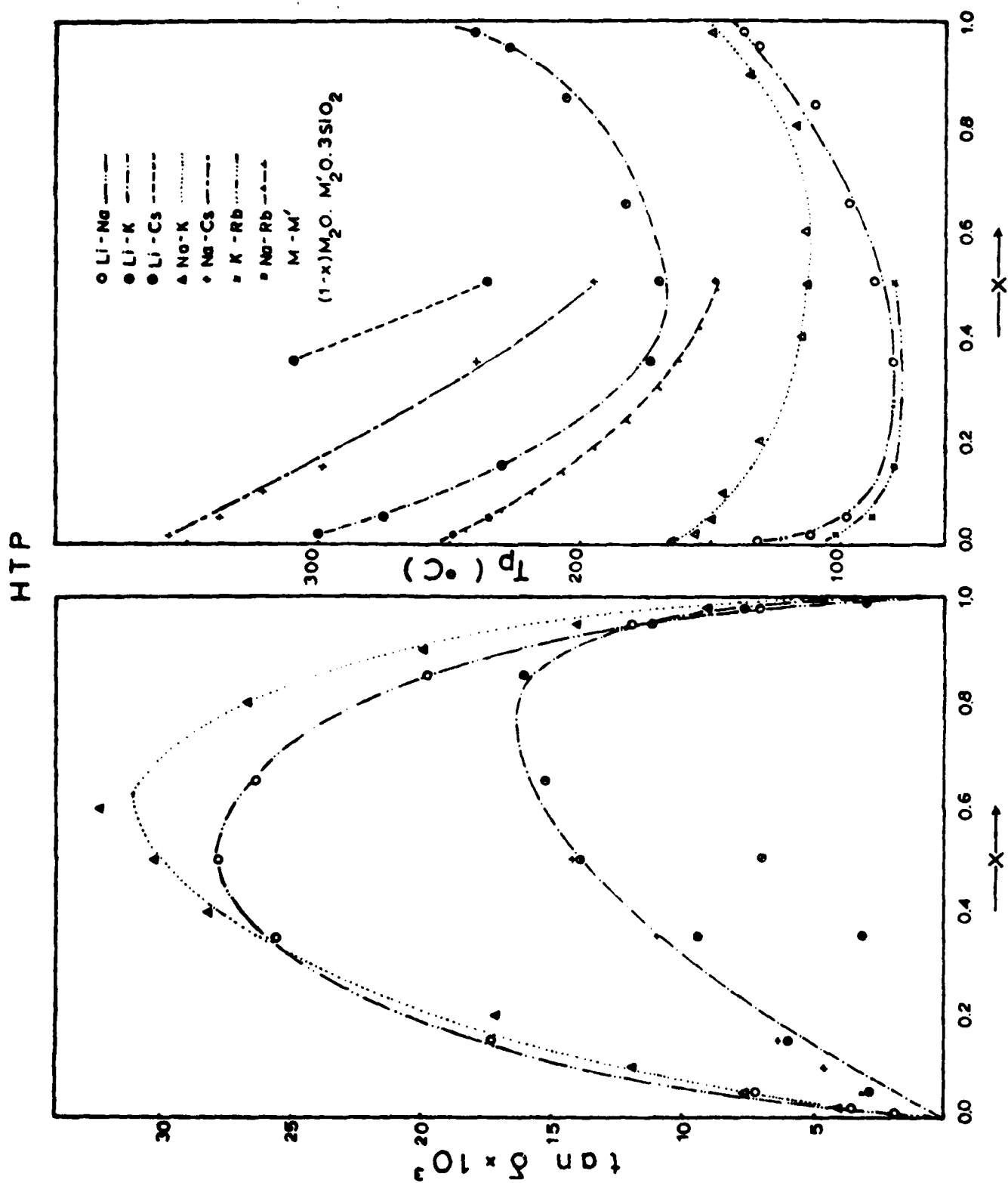
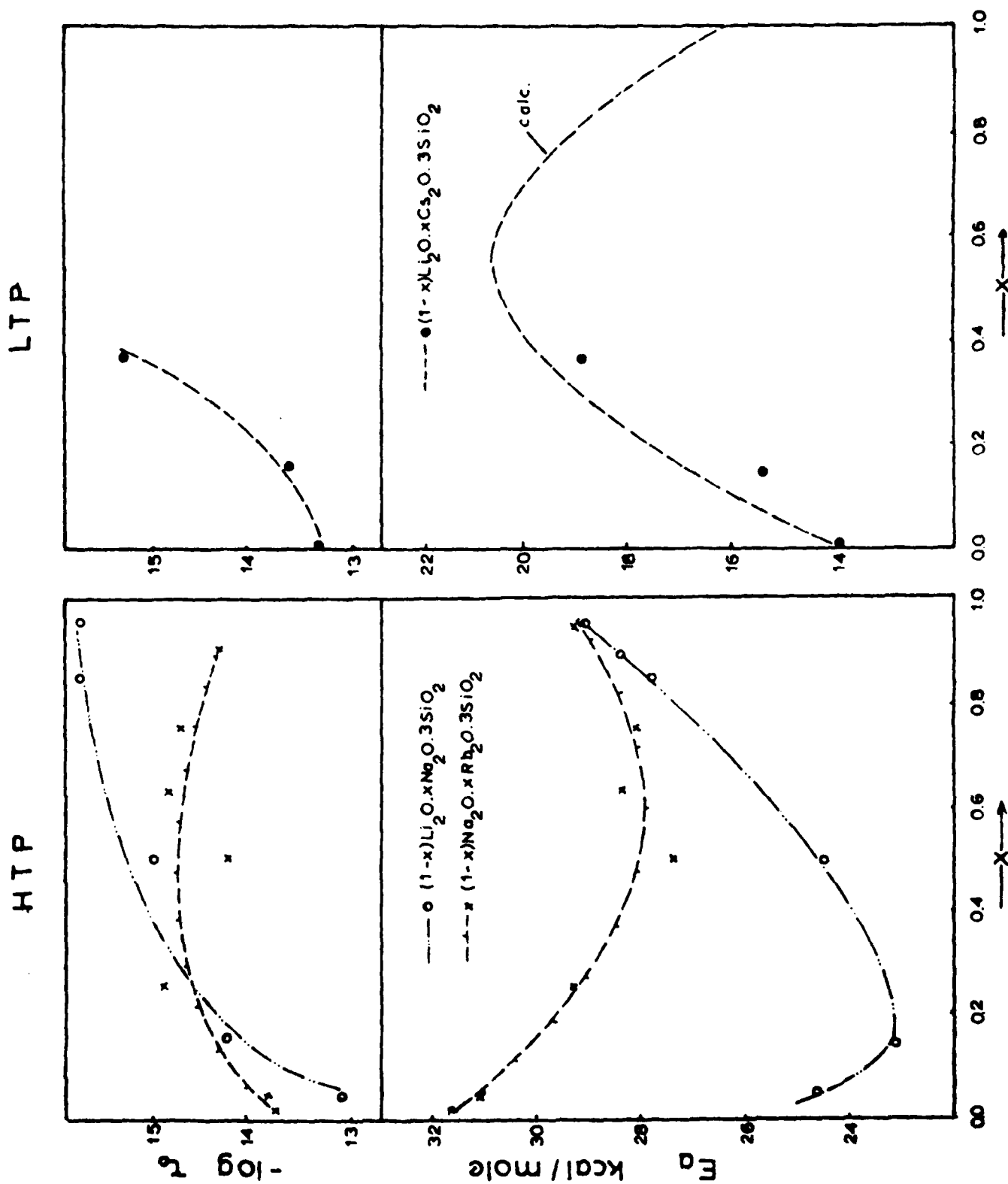


Figure 3

Activation Energies and Preexponential Arrhenius Factors for the HTP and LTP Features of Several Mixed Alkali Glass Systems



of this LTP activation energy $E_a(x)$ correlate well with the behavior of $E_a(x)$ for ionic conductivity (3). Although this could be coincidental, it probably is not since this behavior is characteristic of cooperative cation motion in which all alkali ions participate in a rearrangement. As discussed below it is important to note that this LTP feature is present in the single alkali glasses, so only its composition dependence and not its existence is special to mixed alkali glasses.

The magnitude of $\tan\delta$ for the LTP decreases as the peak temperature, T_p , increases. When the mole fraction x is in the ca. 0.3-0.7 range, the LTP rarely is distinguishable because it becomes buried under the fast-growing HTP, whose peak temperature has decreased to approach that of the LTP. The closer the composition of the mixed alkali glass is to either of the two end numbers, the lower is T_p and the higher is $\tan\delta$. Thus, it is apparent that the LTP $\tan\delta$ decreases from the $\tan\delta$ values of the end member, single alkali glasses, and must have a minimum in the $x = 0-1$ range.

On the other hand, the high temperature peak (HTP) is not clearly apparent in single alkali glasses and has behavior that is entirely different from that of the LTP. Its activation energy, $E_a(x)$, plotted as a function of x in Figure 3, shows a minimum instead of the maximum shown by this function for the LTP. The magnitude of $E_a(x)$ is roughly twice the activation energy for conductivity of the end number single alkali glasses. This clearly indicates that either a new type of high energy barrier is surmounted or that a higher number of low energy barriers must be overcome simultaneously than in the low temperature relaxation. This is more consistent with a network rearrangement involving a larger segment.

The magnitude of $\tan\delta$ for the HTP increases with increasing x until it reaches a maximum and then decreases again as x approaches 1.0. This HTP peak often appears to arise from the background, occasionally is discussed as growing out of

the high temperature loss cut off close to the glass transition, and sometimes appears to be related to a small peak between the LTP and T_g . Whether it is closely related to any of these features depends on the glass, since there can be special features in any system, but it is clear that network rearrangement is a central aspect to the HTP. The main characteristics of the HTP, then, are that it becomes apparent when there are two or more dissimilar cations in the system, has a high activation energy, which varies with x , has a magnitude that rises to a high value and exhibits a maximum in the $x = 0-1.0$ range, and is related to network relaxation.

Toward a Theory

A theory for the mechanical phenomena of mixed alkali glasses must be based on the cooperativity of the motions involved in the activation and relaxation processes. And, it must explain why the two main features of the mechanical loss spectrum behave in opposite ways as the cation composition is varied. Thus, we require a framework within which different cooperative motions give rise to an $E_a(x)$ maximum for the LTP, an $E_a(x)$ minimum for the HTP, a $\tan\delta(x)$ minimum for the LTP, and a $\tan\delta(x)$ maximum for the HTP. This section is designed to discuss ways of approaching the development of a theory for these phenomena.

The materials exhibiting these phenomena can be modelled as ones in which there are two types of cations of the same charge but different size, M_L and M_S (for large and small, +1 charged cations), distributed more or less uniformly in a single phase complex anionic network. Energetic considerations add to this model the fact that the oxygen atoms surrounding the smaller cation will be closer to its nucleus than will those around the larger cation; that is, that they approach a smaller cation more closely and this constitutes a small "site".

The activation free energy of a cooperative process is the difference between the free energies of the cooperativity rearranging and the non-rearranging subsystems of the equilibrium ensemble of the material (3,7). The transition probability for a cooperative rearrangement is given as a function of x by:

$$W(x) = A \exp \frac{-\Delta G^*(x)}{RT} \quad (8)$$

and the associated relaxation time is given by Eqn (5). Thus, the problem of finding $\tau(x)$ involves evaluating $\Delta G^*(x)$. From Eqn (6), this also can be expressed in terms of the entropic and enthalpic activation parameters.

The appropriate composition dependences for ΔG^* depend on the models assumed for the motion involved in the relaxation. Since the materials are complex and the phenomena must be treated statistically, it is not appropriate to assume that a single event is responsible for either relaxation. However, we can begin to treat the problem as follows.

Since the magnitude and composition dependencies of activation energies associated with LTP are nearly the same as those formed for ionic conductivity (3), we assign LTP to a cooperative rearrangement involving dissimilar cations. In this sense, the LTP is actually closer to a mixed alkali peak than is the HTP, although the conventional nomenclature is the opposite. Now employing the regular solution theory (mean field approximation) we have shown (3) that

$$\Delta G^*(x) = x\Delta G_L^* + (1-x)\Delta G_S^* + x(1-x)J \quad (9)$$

where ΔG_L^* and ΔG_S^* are the activation free energies per mole and $x(1-x)J$ a work term associated with the additional energy required by those processes involved in the overall relaxation which involve expansion of a site to accommodate a larger cation. The J term is evaluated from far-infrared spectroscopic data. The $\Delta G^*(x)$ exhibits a maximum in the $0.5 \leq x \leq 1.0$ region. Since it has a maximum, the temperature at which mechanical resonance occurs T_p (LTP) also has a maximum at the same composition.

The HTP in the mechanical loss spectrum must also be treated in a cooperative manner, but its assignment to a network relaxation means that another form of $\Delta G^*(x)$ is more appropriate. The Adam-Gibbs theory (7) expression for the activation free energy associated with cooperative rearrangement of networks is $\Delta G^* = Z^* \Delta \mu$, where Z^* is the critical size (minimum) of a rearranging unit and the free energy barrier per monomer segment is $\Delta \mu$. Employing this, we include the composition dependence and write the relaxation time for a cooperative rearrangement as

$$\tau(x) = \frac{1}{W(x)} = A' \exp \frac{Z^*(x)\Delta\mu(x)}{RT} \quad (10)$$

Spectroscopic evidence (8) shows that the force field around a monomeric segment doesn't change much upon the introduction of a dissimilar cation, at least in mixed alkali glasses in which the network structure itself isn't altered. And, spectral evidence shows (9) that HTP appears even when the introduction of the dissimilar cations do not charge the network. Therefore, to a first approximation $\Delta\mu(x)$ does not have a strong x dependence. Thus, we look to $Z^*(x)$, the critical size of the rearranging unit, to provide the principal contribution to the x dependence of $\Delta G^*(x)$ and thus of T_p (HTP). If $Z^*(x)$ is defined as critical (minimum) in the sense that it represents the smallest rearrangeable subsystem containing a nearly statistical distribution of the two types of cations, several aspects of the interpretation are clarified. First, it focuses on the entropic part of $\Delta G^*(x)$, as it must if $\Delta\mu(x)$ is largely energetic and slowly varying, and indicates that the role of the presence of a distribution of dissimilar cations is to make a large number of configurations available to the rearranging system. This entropic contribution increases to make the rearrangement probable. The critical size $Z^*(x)$ clearly exhibits a minimum with respect to x , allowing $\tau(x)$ and thus T_p (HTP) to exhibit a minimum.

The free energy barrier per monomer does depend on the nature of the cation and network, of course, so the product $Z^*(x)\Delta\mu(x)$ for different pairs of cations will be different for the same composition x .

The magnitudes ($\tan\delta$) of the LTP and HTP, and the details of their composition dependences can also be considered on the basis of these approaches. In the case of the HTP, it is clear that as $Z^*(x)$ decreases, the number of subsystems that can undergo the rearrangement at a given temperature increases.

Therefore, it is appropriate to explore theoretically the fact that the number of subsystems reached a maximum at the same composition that $Z^*(x)$ reaches a minimum, and that $\tan\delta(x)$ reaches a maximum at the same composition that T_p (HTP) reaches a minimum.

On the other hand, the composition dependence of $\tan\delta$ for LTP must be understood in terms of the model used to calculate $\Delta G^*(x)$ for that relaxation. Since the types of collective motion involve sub-events whose energies can be considered somewhat separately and averaged statistically, we may approach the problem by considering the energy losses directly.

A convenient expression for scaling $\tan\delta(x)$ for low energy losses (on the order of 1%) is

$$\tan\delta(x) = \frac{E_{act}(x) - E_{rel}(x)}{E_{act}(x)} \quad (11)$$

where $E_{act}(x)$ represents the energy taken by the rearranging subsystem in being activated, and $E_{rel}(x)$ is the energy returned upon relaxation. Taking account of the number of each type of event occurring as the ensemble rearranges, and setting $\tan\delta^S$ and $\tan\delta^L$ to the loss tangent for rearrangements involving only small or large cations, respectively, it can be shown that

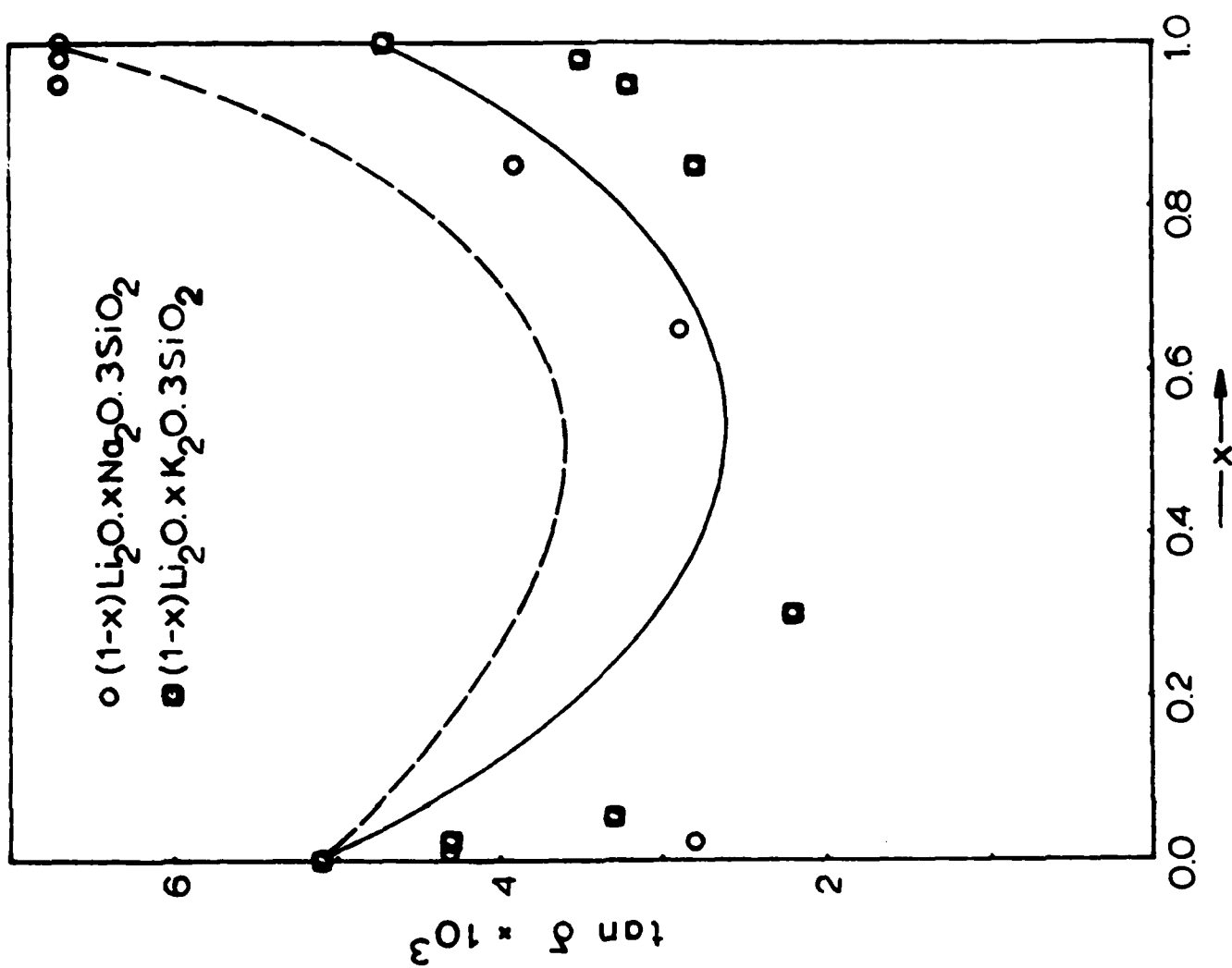
$$\tan\delta(x) = \frac{(1-x)E_a^S \tan\delta^S + x^2 E_a^L \tan\delta^L}{(1-x)E_S + xE_L + x(1-x)W} \quad (12)$$

A comparison of $\tan(x)$ calculated according to Eqn (12) with the experimental data is shown in Figure 4.

In the derivation of Eqn (12) it is necessary to calculate the number of transitions of each type involved in the cooperative rearrangement of the ensemble, and this has been done statistically. However, since these represent the probabilities of cations moving, it is desirable to express this in terms of the cation diffusion coefficients. This can be approached by noting that all cation motion events contribute to the observed diffusion coefficient,

Figure 4

Comparison of the Calculated (Eqn. 12) and Observed Loss Tangent Variations with x for the LTP of Several Mixed Alkali Glass Systems



and establishing the condition that the number of small cations moving in an event is related to the number of large ones moving, by whatever diffusive mechanism, by

$$\frac{n_L}{n_S} = \frac{D^S(x,T)}{D^L(x,T)} \quad (13)$$

where $D^i(x,T)$ are the diffusion coefficients evaluated at the temperature of the event.

Conclusion

In this report the available data on the mechanical behavior of mixed alkali glasses have been analyzed to show that systematic variations (in T_p 's and $\tan\delta$'s) occur and are common to such glasses. The low temperature peak has been reassigned as a mixed alkali peak, rather than one due to previously proposed types of single alkali motion. The high temperature peak has been identified as a network motion, which depends largely on the increased numbers of configurations available during the rearrangement due to the presence of dissimilar cations, rather than its previous identification as the "mixed alkali" peak.

An approach to developing a theoretical understanding of these mechanical features, based on the inherent cooperativity of the relaxations and models suitable to the assignments of the peaks, has been suggested. The details of such a theory are under study and will be reported in a subsequent publication.

Acknowledgment

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